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(54) Title: COMPLIANT AND CROSSLINKABLE THERMAL INTERFACE MATERIALS

(57) Abstract: Described is a compliant and crosslinkable thermal interface material of at least one malenized rubber with maleic anhydride adducted to the molecule and at least one hydroxyl terminated olefin rubbers, and a method of making and using same, as well as a method of improving thermal conductivity of polymer systems.

COMPLIANT AND CROSSLINKABLE THERMAL INTERFACE MATERIALS

Background of The Invention

As electronic devices become smaller and operate at higher speeds, energy emitted in the form of heat increases dramatically. A popular practice in the industry is to use thermal 5 grease, or grease-like materials, alone or on a carrier in such devices to transfer the excess heat dissipated across physical interfaces. Most common types of thermal interface materials are thermal greases, phase change materials, and elastomer tapes. Thermal greases or phase change materials have lower thermal resistance than elastomer tape because of the ability to be spread in very thin layers and provide intimate contact between adjacent surfaces. Typical 10 thermal impedance values range between 0.6-1.6°C cm²/w. However, a serious drawback of thermal grease is that thermal performance deteriorates significantly after thermal cycling, such as from -65C° to 150C°, or after power cycling when used in VLSI chips. It has been also found that the performance of these materials deteriorates when large deviations from 15 surface planarity causes gaps to form between the mating surfaces in the electronic devices or when large gaps between mating surfaces are present for other reasons, such as manufacturing tolerances, etc. When the heat transferability of these materials breaks down, the performance of the electronic device in which they are used is adversely affected. The present invention provides a thermal interface material that is particularly suitable for use as a interface material in electronic devices.

20 Summary of the Invention

In accordance with the invention there is provided a compliant and crosslinkable material which comprises a polymer, such as at least one malenized rubber with maleic anhydride adducted to the molecule and at least one hydroxyl terminated olefin rubber and at 25 least one thermally conductive filler. The compliant thermally conductive material has the capability of enhancing heat dissipation in high power semiconductor devices and maintains stable thermal performance. It is not subject to interfacial delamination or face separation during thermal-mechanical stresses or fluctuating power cycling of the electronic devices in which it is used.

The compliant and crosslinkable thermal interface material may be formulated by 30 mixing the components together to produce a paste which may be applied by dispensing

methods to any particular surface and cured at room temperature or elevated temperature. It can be also formulated as a highly compliant, cured, tacky elastomeric film or sheet for other interface applications where it can be preapplied, for example on heat sinks, or in any other interface situations.

5 The filler to be incorporated advantageously comprises at least one thermally conductive fillers, such as silver, copper, aluminum, and alloys thereof; boron nitride, aluminum nitride, silver coated copper, silver coated aluminum and carbon fiber. It may be also additionally useful to incorporate antioxidants to reduce oxidation of the rubbers, wetability enhancing agents to promote wetting of surfaces, curing accelerators, such as
10 10 would allow curing at room temperature, viscosity reducing agents to enhance dispersability and crosslinking aids. It is also often desirable to include substantially spherical particles of filler to limit the compressibility of the compliant material in interface applications, i.e. to limit or control the thickness.

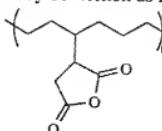
15 It has been also found that thermal conductivity of polymer-resin filler systems, such as a combination of filler and the combined malenized rubber and olefin rubber discussed above, can be especially improved by incorporating carbon micro fibers, with other fillers, into the system.

Detailed Description

20 A compliant and crosslinkable thermal interface material is formulated by combining at least one saturated or unsaturated malenized rubber with maleic anhydride adducted the molecule and at least one saturated or unsaturated hydroxyl terminated olefin rubber and at least one thermally conductive filler. More than one rubber of each type may be combined to produce a compliant and crosslinkable interface material. Olefin containing interface materials, with appropriate thermal fillers, exhibit a thermal capability of less than $0.5 \text{ cm}^2\text{K}$
25 c/w. Unlike thermal grease, thermal performance of the material will not degrade after thermal cycling or flow cycling in IC devices because liquid olefins will cross link to form a soft gel upon heat activation. Moreover, when applied as an interface material it will not be "squeezed out" as thermal grease does in use and will not display interfacial delamination during thermal cycling. The new material can be provided as a dispensable liquid paste to be
30 applied by dispensing methods and then cured as desired. It can also be provided as a highly

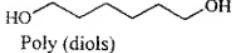
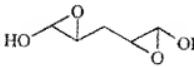
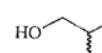
compliant, cured, elastomer film or sheet for pre-application on interface surfaces, such as heat sinks. Advantageously, fillers with a thermal conductivity of greater than about 0.2, and preferably at least about 0.4, w/m°C will be used. Optimally, it is desired to have a filler of not less than about 1 w/m°C thermal conductivity. The compliant thermally conductive

5 material enhances thermal dissipation of high power semiconductor devices. The paste may be formulated as a mixture of functional liquid rubbers and thermal fillers. The functional liquid rubbers are malenized liquid rubbers, saturated or unsaturated, which contain maleic anhydride adducted to the molecule. For example, malenized polybutadiene (commercially available by Ricon Resins, Inc.), malenized poly (styrene-butadiene), etc. The formula for 10 malenized liquid rubber may be written as follows:



Malenized liquid rubber

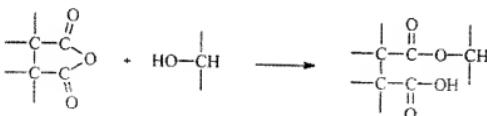
Hydroxyl terminated liquid olefin rubbers, saturated or

15 HO  Poly (diols) HO  HO  (Triol)

unsaturated, useful in accordance with the invention include, by way of example, hydroxyl terminated polybutadiene, hydroxyl terminated epoxidized polybutadiene, such as available from Elf Atochem of Philadelphia, Pennsylvania, hydroxyl terminated hydrogenated polyisoprene, such as available from Kuraray of Japan, hydroxyl functional, poly

20 (ethylene/butlenes) polymers, such as Kraton Liquid available from Shell Chemical. The formula for the hydroxyl terminated liquid olefin rubbers may be written as follows:

Trifunctional polydiols, as fatty ether triol might be used for additional crosslinking. The anhydride functional group reacts with the hydroxyl group as follows:



The anhydride functional group can also react with an epoxide to yield an ester linkage. The reaction between the two functional liquid rubbers can occur at temperature about 50 to 150° C to form a crosslink elastomer network. A tertiary amine may be added as an accelerator which would allow to cure at room temperature. The concentration of maleic

5 anhydride determine the crosslinking density of the elastomer. Physical properties can be varied from a very soft gel material at a very low crosslink density to tough elastomer network of higher crosslink density. The anhydride ring can react with hydroxyls, amines, epoxies to form an elastomer network. Typical liquid rubber mixtures have a viscosity of about 10-200 poises at 25° C.

10 Thermal filler particles to be dispersed in the liquid rubber mixture should advantageously have a high thermal conductivity. Suitable filler materials include silver, copper, aluminum, and alloys thereof; boron nitride, aluminum nitride, silver coated copper, silver coated aluminum and carbon fibers. Combinations of boron nitride and silver or boron nitride and silver/copper also provide enhanced thermal conductivity. Boron nitride in 15 amounts of at least 20 wt. % and silver in amounts of at least about 60 wt. % are particularly useful.

Of special efficacy is a filler comprising a particular form of carbon fiber referred to as "vapor grown carbon fiber" (VGCF) such as is available from Applied Sciences, Inc., Cedarville, Ohio. VGCF, or "carbon micro fibers", are a highly graphitized type by heat 20 treatment (thermal conductivity = 1900 w/m°C). Addition of about 0.5 wt. % carbon micro fibers provides significantly increased thermal conductivity. Such fibers are available in varying lengths and diameters; namely, 1 mm to tens of centimeters in length and from under 0.1 to over 100 μm in diameter. One useful form has a diameter of not greater than about 1 μm and a length of about 50 to 100 μm , and possess a thermal conductivity of about two or 25 three times greater than with other common carbon fibers having diameters greater than 5 μm .

It is difficult to incorporate large amounts of VGCF in polymer systems such as the malenized rubber and olefin rubber combination discussed above. When carbon micro fibers, e.g. (about 1 μm , or less, are added to the polymer they do not mix well because the need to incorporate a large amount of fiber relative to the amount of polymer for beneficial 30 improvement thermal conductivity. However, we have found that relative large amounts of

carbon micro fibers can be added to polymer systems that have relatively large amounts of other fillers. A greater amount of carbon micro fibers can be added to the polymer when added with other fibers than can be added alone to the polymer, thus providing a greater benefit with respect to improving thermal conductivity of the thermal interface material.

5 Desirably, the ratio of carbon micro fibers to polymer is in the range of 0.05 to 0.50 by weight.

It is also advantageous to incorporate substantially spherical filler particles to maximize packing density. Additionally, substantially spherical shapes or the like will also provide some control of the thickness during compaction. Dispersion of filler particles can be
10 facilitated by addition of functional organo metallic coupling agent such as organosilane, organotitanate, organozirconium, etc. Typical particle sizes useful for fillers in the rubber material may be in the range of about 1-20 μm with a maximum of about 100 μm . Antioxidants may be added to inhibit oxidation and thermal degradation of the cured rubber
gel. Typical useful antioxidants include Irganox 1076, a phenol type of Irganox 565, an
15 amine type, (at 0.01% to about 1 wt.%), available from Ciba Giegy of Hawthorne, New York. Typical cure accelerators include tertiary amines such as didecyldiethylamine, (at 50 ppm - 0.5 wt. %)

To illustrate the invention, a number of examples were prepared by mixing the components described in Examples A through R below. For these examples reference is
20 made to the commercial trade designation which is defined at the conclusion of the tables describing the samples.

As indicated in the tables, the properties of the compositions of viscosity, pot life, appearance after curing, modulus, glass transition temperature and thermal conductivity are also reported.

25 The examples shown include one or more of the optional additions, e.g., antioxidant, wetability enhancer, curing accelerators, viscosity reducing agents and crosslinking aids. The amounts of such additions may vary but, generally, they may be usefully present in the following approximate amounts (in wt. %): filler up to 95% of total (filler plus rubbers); wetability enhancer 0.1 to 1% (of total); antioxidant 0.01 to 1% (of total); curing accelerator
30 50 ppm - 0.5% (of total); viscosity reducing agents 0.2 - 15%; and crosslinking aids 0.1-2%.

It should be noted the addition at least about 0.5% carbon fiber significantly increases thermal conductivity.

TABLE 1
Compositions With Thermal Fillers

	1	2	3	4	5	6	7
RICON 130 MA8	5	5	5	5	5	25	16
PolyBd R45	5	5	5			25	16
Kapton L2203				5			
TH21					5		
Ag Flake 1	90						
Ag Flake 2		90					
Ag Cu 107			90	90	90		
Boron Nitride PT120						50	
Aluminum Nitride							68
Viscosity (PaS)	100	100	150	200	180	210	200
Pot Life @ 25°C	<4 hrs.	>12 hrs.	>12 hrs.	>12 hrs.	>12 hrs.	>12 hrs.	>12 hrs.
Appearance after Cured @ 150°C/30 min.	N/A	----(flexible elastomer film)----					
Modulus @ 25°C	N/A	1.2	1.2	1.4	1.1	0.8	1.1
Glass Transition Temp (°C)	N/A	-80	-80	-65	-75	-80	-75
Thermal Conductivity w/m°C	N/A	1.5	1.45	1.3	1.4	1.0	1.0

TABLE 2
Compositions With Varying Cross-link Densities

	8	9	10	11	12	13
RICON 130 MA8	3.76	1.88	0.64	1.13	5.64	6.27
PolyBd R45	3.76	5.64	6.88	6.39	1.88	1.25
KR-TTS	0.40	0.40	0.40	0.40	0.40	0.40
Irganox 565	0.04	0.04	0.04	0.04	0.04	0.04
Irganox 1032	0.04	0.04	0.04	0.04	0.04	0.04
Ag Flake 2	91.5	91.5	91.5	91.5	91.5	91.5
Ag Sphere	0.5	0.5	0.5	0.5	0.5	0.5
Viscosity (PaS)	170	160	151	155	160	160
Appearance after Cured @ 150°C/30°	non-tacky flexible film	tacky film	tacky soft gel	tacky film	flexible film	flexible film
Modulus @ RT (Mpa)	1.3	0.7	0.2	0.4	1.0	1.5
Thermal Conductivity w/m°C	1.5	1.8	1.9	1.8	1.9	1.8

Note: Ag spheres, 50 μ m diameter are added spacers for bond thickness control.

TABLE 3
Room Temperature Curable Compositions

	14	15
DAMA 1010 Amine	0.1	0.15
RICON 130 MA8	1.1	1.07
PolyBd R45	6.32	6.30
KRTTS	0.40	0.40
Irganox 565	0.04	0.04
Irganox 1032	0.04	0.04
Ag Flake 2	91.5	91.5
Ag Sphere	0.5	0.5
Viscosity (cps)	155	160
Appearance after Cure	tacky film	tacky film
Modulus @ RT (Mpa)	0.4	0.4
Thermal Conductivity w/m°C	1.9	1.9

TABLE 4
 Compositions with Mixed Thermal Fillers,
 Reactive Diluents and Additional Cross-linker

	16	17	18
KRTTS	0.4	0.42	0.81
RICON 130 MA8	0.90	1.0	1.94
PolyBd R45	5.00	5.63	10.91
Drakeol 9LT	0.52	0.75	1.45
Sovernol VP 95	0.1	0.10	0.19
Irganox 1076	0.04	0.05	0.1
Irganox 565	0.04	0.05	0.1
Ag Flake 2	91.5		
Ag Cu 107		89.5	80
Boron Nitride PT 120	1.0	2	4
Ag Sphere	0.5	0.5	0.5
Viscosity (cps)	114	120	70
Cure @: 150°C/30 min		-----tacky film-----	
Modulus @ RT (Mpa)	0.5	0.4	0.3
Thermal Conductivity w/m°C	2.5	3.0	1.5

TABLE 4 (Cont'd)

	19	20	21	22	23	24	Δ
KRTTS	0.4	0.4	0.4	2.8	1.25	1.0	5
RICON 130 MA8	5.0	5.0	5.0	35.7	17.9	13.2	71.5
PolyBd R45	1.0	1.0	1.0	7.2	3.6	2.9	14.5
Drakeol 9LT	0.42	0.42	0.42	3.0	1.6	1.3	6.5
Sovernol VP95	0.1	0.1	0.1	0.7	0.35	0.26	1.3
Irganox 1076	0.04	0.04	0.04	0.3	0.15	0.12	0.6
Irganox 565	0.04	0.04	0.04	0.3	0.15	0.12	0.6
Ag Flake	91.5	91.5	91.5				
Ag Sphere	0.5	0.5	0.5				
Boron Nitride (PT120)	1.0	--	--	45			
Carbon Fiber (VGCF)	--	1.0	2.0	5	5	5	
Silica Powder	--	--	--	--	70	--	
Aluminum Powder	--	--	--	--	--	75	
Viscosity (kcps)	125	140	175	130	140	120	3.2
Thermal Conductivity w/m°C	2.5	3.5	4.1	1.8	1.4	3.0	0.2
Thermal Impedance cm ² °C/W	0.42	0.24	0.20	0.45	0.6	0.40	4.8
Carbon Fiber/Resin/ration	0	0.14	0.20	0.10	0.2	0.25	

DEFINITIONS:

RICON 130 MA8: Polybutadiene, molecular weight 3100, adducted with 8% maleic anhydride per rubber weight supplied by Ricon Resins, Inc. of Grand Junction, Colorado.

5 Poly Rd R45: Hydroxyl terminated polybutadiene, molecular weight ~ 2700 supplied by Elf Atochem of Philadelphia, Pennsylvania.

10 DAMA 1010 Amine: Didecylmethylamine which is a tertiary amine acting as cure accelerator supplied by Albemarle Corp. of Baton Rouge, Louisiana.

15 Drakeol 9LT: Mineral oil as reactive diluent supplied by Pennreco of Dickerson, Texas.

20 Kapton L2203: Hydroxyl terminated poly (Ethylene/Butylene), molecular weight ~ 4000 supplied by Shell Chemical of Houston Texas.

VGCF fiber: A high graphitized type by heat treatment (Thermal conductivity = 1900 w/m C supplied by Applied Sciences of Cedarville, Ohio.

25 TH21: Hydroxyl terminated hydrogenated Polyisoprene (supplied by Kuraray, Japan).

Ag Flake 1: Contains an amino type of surfactant coating which acts as accelerator causing shortened pot life.

25 Ag Flake 2: Contains a surfactant coating inert to the cure reaction resulting in longer pot life.

30 Ag Cu 107: Silver plated copper powder.

KR-TTS: Organotitanate coupling agent added to enhance dispersion of fillers supplied by Kenrich of Bayonne, New Jersey.

35 Sovernol VP95: Fatty ether triol as additional crosslinker, supplied by Henkel Corp. of Kankakee, Illinois.

Irganox 565 & 1076: Antioxidants supplied by Ciba Geigy of Hawthorne, New York.

40 It is apparent from the foregoing that various changes and modifications may be made without departing from the invention. For example, more than one form of rubber of each type, or other additive, may be combined and more than one type of filler may be included. It is also clear by comparing the properties of all filler-containing examples with Example A of Table 4 that the combination of rubbers without filler has significantly less

thermal conductivity, and greater thermal impedance, than the filler-containing examples. It is also evident the thermal conductivity of polymers, such as the rubbers described, can be improved most by incorporating carbon micro fibers. Accordingly, the scope of the invention should be limited only by the appended claims, wherein what is claimed is:

1. A compliant and crosslinkable material useful as an interface material for electronic devices comprising at least one saturated or unsaturated malenized rubber with maleic anhydride adducted to the molecule, at least one saturated or unsaturated hydroxyl terminated olefin rubber and at least one thermally conductive filler.
- 5 2. A compliant and crosslinkable material according to claim 1 comprising about 8 to 92 wt.% hydroxyl terminated olefin rubber.
3. A compliant and crosslinkable material according to claim 1 wherein said filler comprises at least one of silver, copper, aluminum, and alloys thereof; boron nitride, aluminum nitride, silver coated copper, silver coated aluminum, and carbon fibers; and 10 mixtures thereof.
4. A compliant and crosslinkable material according to claim 3 further comprising carbon micro fibers in addition to at least one other filler.
5. A compliant and crosslinkable material according to claim 1 wherein said filler comprises said carbon micro fibers and at least one other filler comprising silver, copper, 15 aluminum, and alloys thereof; boron nitride, aluminum nitride, silver coated copper, silver coated aluminum, and carbon fibers; and mixtures thereof.
6. A compliant and crosslinkable material according to claim 1 wherein at least one portion of said filler comprises substantially spherical particles.
7. A compliant and crosslinkable material according to claim 1 further comprising an 20 antioxidant.
8. A compliant and crosslinkable material according to claim 7 wherein said antioxidant is present in an amount of 0.01 to 1 wt. %.
9. A compliant and crosslinkable material according to claim 1 further comprising a coupling agent.
- 25 10. A compliant and crosslinkable material according to claim 9 wherein said coupling agent is present in an amount of about 0.1 to 2 wt.%.

11. A compliant and crosslinkable material according to claim 1 further comprising a curing accelerator.
12. A compliant and crosslinkable material according to claim 11 wherein said curing accelerator is present in an amount of about 50 ppm to 0.5 wt.%.
- 5 13. A compliant and crosslinkable material according to claim 1 further comprising a wetability enhancer.
14. A compliant and crosslinkable material according to claim 13 wherein said wetability enhancer is present in an amount of 0.1 to 2 wt.%.
- 10 15. A compliant and crosslinkable material according to claim 1 further comprising a viscosity adjusting agent.
16. A compliant and crosslinkable material according to claim 15 wherein said viscosity adjusting agent is present in amount of about 0.2 to 15 wt.%.
17. A dispensable paste of a compliant and crosslinkable material useful as an interface material for electronic devices comprising at least one malenized rubber with maleic anhydride adducted to the molecule and at least one hydroxyl terminated olefin rubber and at least one thermally conductive filler.
- 15 20 18. A sheet or film of a compliant and crosslinkable material useful as an interface material for electronic devices comprising at least one malenized rubber with maleic anhydride adducted to the molecule, at least one hydroxyl terminated olefin rubber and at least one thermally conductive filler.
19. A method of making a compliant and crosslinkable material comprising combining at least one malenized rubber with maleic anhydride adducted to the molecule, at least one hydroxyl terminated olefin rubber and at least one thermally conductive filler.
20. A method according to claim 19 further comprising formulating a dispensable paste of said material.

21. A method according to claim 19 further comprising molding said material as a sheet or film capable of being cut to size and applied as an interface between components in an electronic device.
22. A compliant and crosslinkable material according to claim 1 comprising up to 95 wt.% filler, said filler comprising carbon micro fibers and at least one of silver, copper, aluminum, and alloys thereof; boron nitride, aluminum nitride, silver coated copper, silver coated aluminum, and carbon fibers; and mixtures thereof.
23. A compliant and crosslinkable material according to claim 1 wherein said filler comprises about 90 to 95 wt.% of the material.
- 10 24. A method according to claim 19 further comprising incorporating a filler comprising at least one of silver, copper, aluminum, and alloys thereof; boron nitride, aluminum nitride, silver coated copper, silver coated aluminum and carbon fibers; and mixtures thereof.
25. A method according to claim 24 further including carbon micro fibers in addition to one or more other fillers.
- 15 26. A method of increasing the thermal conductivity of a polymer comprising incorporating therein carbon micro fibers and at least one thermally conductive filler.
27. A method according to claim 26 wherein said carbon micro fiber is present in an amount of at least about 0.5 wt.%, or in a ratio of carbon micro fibers to polymer of at least 0.05.
- 20 28. A method according to claim 27 further comprising additionally incorporating a filler comprising at least one of silver, copper, aluminum, and alloys thereof; boron nitride, aluminum nitride, silver coated copper, silver coated aluminum, and carbon fibers; and mixtures thereof.
29. A method according to claim 26 wherein said polymer comprises at least one saturated or unsaturated malenized rubber with maleic anhydride adducted to the molecule and at least one saturated or unsaturated hydroxy terminated olefin rubber.

INTERNATIONAL SEARCH REPORT

International application No.
PCT/US00/25447

A. CLASSIFICATION OF SUBJECT MATTER

IPC(7): H01B 1/02, 1/04, 1/06; C08L 25/04, 33/02; C08K 3/00;

US CL. 252/503, 511, 512, 514; 524/439, 910; 525/74, 78, 207, 221, 285;

According to International Patent Classification (IPC) or to both national classification and IPC

B. FIELDS SEARCHED

Minimum documentation searched (classification system followed by classification symbols)

U.S. : 252/503, 511, 512, 514; 524/439, 910, 525/74, 78, 207, 221, 285;

Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched

Electronic data base consulted during the international search (name of data base and, where practicable, search terms used)

C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
X	US 5,989,459 A [NGUYEN et al] 23 November 1999, entire document, especially claims 1-25.	1-25
A	US 5,300,569 A [DRAKE et al] 05 April 1994, entire document.	1-29
Y,P	US 5,990,217 A [NAKANO et al] 23 November 1999, col. 5, line 5 - col. 6, line 1.	26-29

 Further documents are listed in the continuation of Box C. See patent family annex.

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